

Magnetic anisotropy and reversal in epitaxial Fe/MgO(001) films revisited

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We investigate the magnetization reversal in Fe/MgO(001) films with fourfold in-plane magnetic anisotropy and an additional uniaxial anisotropy whose orientation and strength are tuned using different growth geometries and post growth treatments. The previously adopted mechanism of 180° domain wall nucleation clearly fails to explain the observed 180° magnetization reversal. A new reversal mechanism with two successive domain wall nucleations consistently predicts the switching fields for all field orientations. Our results are relevant for a correct interpretation of magnetization reversal in many other epitaxial metallic and semiconducting thin films.

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Magnetic anisotropy is one of the most important properties of metallic and semiconducting thin-film magnets [1, 2]. In magnetic films of cubic systems an in-plane fourfold magnetic anisotropy is expected, but often an additional uniaxial magnetic anisotropy (UMA) is observed to be superimposed on top of the fourfold anisotropy [3, 4]. The extra UMA has been attributed to different origins, including a self-shadowing effect occurring during oblique deposition [5, 6, 7], the bonding between film and substrate [8, 9], and the Néel surface effect on a stepped substrate [10, 11]. Moreover, ion sputtering has been demonstrated as a reliable tool to control the orientation and strength of UMA [12, 13].

When applying thin films for magnetic data storage and spintronic devices, the magnetization reversal mechanisms and their dependence on the anisotropy symmetry need to be known and controlled. The magnetization reversal process for combined cubic and uniaxial anisotropies is sensitive to the specific anisotropy geometry and strength [14, 15]. Depending on the field orientation, hysteresis curves with one and two steps are observed in various films, and explained in terms of nucleation and propagation of 90° and 180° domain walls (DWs) [16, 17]. A model based on minimizing the magnetic energies has been introduced with DW nucleation energies ϵ_{90° for 90° DWs and ϵ_{180° for 180° DWs, respectively, in order to account for the observed switching fields [17]. A special magnetic switching process involving three steps can be observed when the additional UMA along the cubic easy axis exceeds ϵ_{90° [18, 19]. Until now, such a switching has been assumed to be mediated by two 90° DW nucleations at the first and the third step and one 180° DW nucleation occurring in between.

Previous work on Fe/MgO(001) films grown at normal incidence revealed a weak UMA along Fe(010) [20]. Recently, we successfully relied on ion sputtering to manipulate the strength of the in-plane UMA along Fe(110) in Fe/MgO(001) [21]. Park *et al.* found that a pronounced UMA can be induced in Fe/MgO(001) by rely-

ing on oblique-incidence molecular beam epitaxy (MBE) growth [22]. Up to now, all of the measured hysteresis loops in Fe/MgO(001) only revealed one or two steps.

Here, we report on a detailed study of magnetization reversal in Fe/MgO(001) films, where strength and orientation of UMA are tuned either by ion sputtering or by oblique-incidence MBE growth. A novel mechanism is introduced with two successive DW nucleations to explain the 180° magnetic switching that occurs for one-step and three-step loops. Our model consistently explains the experimental results for films with different UMA, revealing the universal nature of the magnetization reversal.

In general, the in-plane UMA, which is superimposed on the cubic anisotropy K_1 of Fe, can be separated into two components: K_{u1} along [010] and K_{u2} along [110] [23]. If $K_{u1} \ll K_1$ and $K_{u2} < K_1$, the component K_{u2} rotates the position of the overall easy axes backwards with respect to the uniaxial hard axis over an angle δ that is approximately given by $\delta = \frac{1}{2} \sin^{-1}(K_{u2}/K_1)$ [15], as illustrated in Fig. 1(a). In order to obtain a UMA with different orientation and strength, three Fe films were grown by MBE on MgO(001) substrates using different growth conditions and post growth treatments. For sample A, the incident Fe beam was at an angle of 49° with respect to the surface normal and with azimuthal angle along Fe[010]. During deposition of sample B at normal incidence, the substrate was rotated around the surface normal. The nominal thickness of samples A and B was 15 nm, as monitored by a calibrated quartz crystal oscillator. The growth geometry for sample C was the same as for sample B, but the nominal thickness was 100 nm. Subsequently, sample C was sputtered with 2 keV Ar⁺ ions at an incidence angle of 60° with respect to the surface normal and with azimuth fixed in between Fe[100] and Fe[110]. After sputtering for 250 minutes, the film thickness was reduced to 15 nm, as verified by *ex situ* x-ray reflectometry. Before removing the samples from the vacuum chamber, they were capped with a 2 nm thick protective Au layer. The magnetic

properties were measured by the longitudinal and transverse magneto-optical Kerr effect (MOKE) for different field orientation ϕ as defined in Fig. 1(a).

For sample A, a considerable UMA along $[010]$ is introduced by the oblique deposition. Three-step loops as well as one-step and two-step loops are observed at different ϕ , as illustrated in Figs. 1(b) to 1(d). The switching events, which occur for increasing field and $0^\circ < \phi < 90^\circ$, are $[\bar{1}00] \rightarrow [100]$ for the one-step loops, $[0\bar{1}0] \rightarrow [\bar{1}00] \rightarrow [010]$ for the two-step loops, and $[010] \rightarrow [\bar{1}00] \rightarrow [100] \rightarrow [010]$ for the three-step loops [19]. The magnetization switches by 180° for the one-step loops and for the second step of the three-step loops, and by 90° for the other steps. The corresponding spin orientations are marked by the arrows that are enclosed in a square in Figs. 1(b) to 1(d).

Up to now 90° as well as 180° DW nucleations have been invoked to interpret the 90° and 180° magnetic transitions, respectively [17, 19]. The coercivity related to the DW nucleation energy can be derived from the energy gain between the local minima at the initial and final easy axes involved in the transition [17]. The theoretical switching fields are obtained as $H_{c1} = (\epsilon_{90^\circ} - K_{u1})/[M(\sin \phi - \cos \phi)]$ for the magnetic switching process $[0\bar{1}0] \rightarrow [\bar{1}00]$, $H_{c2} = (\epsilon_{90^\circ} + K_{u1})/[M(\sin \phi + \cos \phi)]$ for $[\bar{1}00] \rightarrow [010]$, $H_{c3} = (\epsilon_{90^\circ} + K_{u1})/[M(\sin \phi - \cos \phi)]$ for $[100] \rightarrow [010]$, $H_{c4} = (\epsilon_{90^\circ} - K_{u1})/[M(\cos \phi - \sin \phi)]$ for $[010] \rightarrow [100]$, and $H_c = \epsilon_{180^\circ}/[2M(\cos \phi)]$ for $[\bar{1}00] \rightarrow [100]$, where M is the magnetization.

For sample A, the ϕ dependence of the measured switching fields can be nicely fitted using the theoretical switching fields (see Fig. 2(a)), provided we assume the switching fields correspond to H_{c1} , H_{c2} and H_{c3} (see Figs. 1(c) and 1(d)). The fitting results in the parameters $K_{u1}/M = 2.70 \pm 0.02$ mT and $\epsilon_{90^\circ}/M = 0.61 \pm 0.02$ mT, where $K_{u1} > \epsilon_{90^\circ}$ is the necessary condition for the occurrence of three-step loops [19]. Following Refs. [17, 19] we try to describe the experimental switching field H_{c180° , which corresponds to a 180° magnetic transition, in terms of 180° DW nucleation. Using the corresponding theoretical switching field H_c , we obtain the fit that is shown in the two insets to Fig. 2(a). H_c reaches a minimum at $\phi = 0^\circ$ and a maximum at $\phi = 90^\circ$. The experimental switching field H_{c180° reveals, however, a peak at both $\phi = 0^\circ$ and 90° . Moreover, around $\phi = 90^\circ$ H_c has a slope that disagrees with experiment. 180° DW nucleation clearly fails to describe the 180° magnetic transition in sample A. Surprisingly, the theoretical expression for H_{c2} , which corresponds to a 90° DW nucleation, allows us to fit the H_{c180° data (see below for more details).

The universal character of the absence of 180° DW nucleation for Fe/MgO(001) is confirmed by the ϕ dependence of the switching fields for samples B and C in Figs. 2(b) and 2(c). Only one-step and two-step loops are observed. As discussed in our recent publication [21], the

switching route for the two-step loops appearing in samples B and C ($180^\circ - \delta \rightarrow 90^\circ + \delta \rightarrow -\delta$ for $-\delta < \phi < 45^\circ$) is different from the path for sample A. When $\delta = 0$, i.e., $K_{u2} = 0$, the experimental switching fields for this type of two-step loop should correspond to the above derived expressions for H_{c2} and H_{c4} . In case $\delta \neq 0$, i.e., $K_{u2} \neq 0$, the theoretical expressions need to be extended:

$$H_{c2} = \frac{\epsilon_{90^\circ - 2\delta} + K_{u1}(\cos^2 \delta - \sin^2 \delta)}{M[\cos(\phi + \delta) + \sin(\phi - \delta)]},$$

$$H_{c4} = \frac{\epsilon_{90^\circ + 2\delta} - K_{u1}(\cos^2 \delta - \sin^2 \delta)}{M[\cos(\phi + \delta) - \sin(\phi - \delta)]},$$

where $\epsilon_{90^\circ - 2\delta}$ and $\epsilon_{90^\circ + 2\delta}$ are the corresponding DW nucleation energies.

For sample B the two switching fields (H_{c2} , H_{c4}) have a dependence on ϕ that is symmetric about $\langle 100 \rangle$. Moreover, the angular dependence of H_{c2} reveals a clear, abrupt step when crossing $\langle 110 \rangle$, as illustrated in Fig. 2(b). We conclude that sample B has a small in-plane UMA along $[010]$ [17]. The fitting parameters are $K_{u1}/M = 0.19 \pm 0.01$ mT and $\epsilon_{90^\circ}/M = 0.36 \pm 0.01$ mT. Because $K_{u1} < \epsilon_{90^\circ}$, three-step loops cannot be observed.

In sample C, UMA with components along both $[010]$ and $[110]$ is introduced by the Ar^+ ion sputtering. The overall easy axes are observed to deviate from $\langle 100 \rangle$ by an angle $\delta = 3^\circ$, i.e., $K_{u2}/K_1 \approx 0.1$. From the results in Fig. 2(c) we find that the UMA component along $[010]$ and the DW nucleation energies are $K_{u1}/M = 1.69 \pm 0.02$ mT, $\epsilon_{90^\circ - 2\delta}/M = 1.83 \pm 0.02$ mT, and $\epsilon_{90^\circ + 2\delta}/M = 2.29 \pm 0.02$ mT, respectively. Because K_{u1} is comparable to $\epsilon_{90^\circ - 2\delta}$, the path $270^\circ + \delta \rightarrow -\delta \rightarrow 90^\circ + \delta$ is energetically more favorable when compared to the counter-clockwise path via $180^\circ - \delta$ for the whole range of angles $45^\circ < \phi < 135^\circ$. Consequently, both H_{c2} and H_{c4} change monotonously within this range.

We again try to describe H_{c180° for the one-step loops of samples B and C in terms of 180° DW nucleation, as illustrated in the insets to Figs. 2(b) and 2(c), respectively. The theoretical curves clearly disagree with experiment. From the fitting for our samples with different anisotropy geometry and strength we conclude that $90^\circ \pm 2\delta$ magnetic transitions in Fe/MgO(001) films are mediated by $90^\circ \pm 2\delta$ DW nucleation, but 180° magnetization reorientations are not mediated by 180° DW nucleation.

Which mechanism dominates the 180° magnetic reversal? Based on the obtained values for K_{u1} and ϵ_{90° , we plot the energy difference between the relevant easy axes as a function of the applied field in Fig. 3(a) for sample B at $\phi = 10^\circ$ and in Fig. 3(b) for sample A at $\phi = 65^\circ$, respectively. In the previously adopted model, the energy difference between 180° and 0° is treated in terms of *one single barrier*, and 180° DW nucleation occurs at H_c when $\Delta E_{180^\circ \rightarrow 0^\circ} = \epsilon_{180^\circ}$, where ϵ_{180° is assumed to correspond to $2\epsilon_{90^\circ}$ [19]. According to our analysis

the switching between $[\bar{1}00]$ and $[100]$ is governed by *two separate energy barriers* between $[\bar{1}00]$ and $[010]$ and between $[010]$ and $[100]$, respectively. The switching then corresponds to two 90° DW nucleation processes. The energy barrier for the transition from $[\bar{1}00]$ and $[010]$ becomes $\Delta E_{180^\circ \rightarrow 90^\circ} = \epsilon_{90^\circ}$ at H_{c2} . However, since $\Delta E_{90^\circ \rightarrow 0^\circ}$ already exceeds ϵ_{90° at H_{c2} , the domains along $[010]$ are unstable and cannot grow. Therefore, a second nucleation of domains along the final $[100]$ remanent direction occurs at H_{c2} , and the two successive 90° DW nucleations appear as one single step in the MOKE loops. In case $\delta \neq 0$, this process consists of a $90^\circ - 2\delta$ DW nucleation and a subsequent $90^\circ + 2\delta$ DW nucleation, or vice versa. Based on our new model the experimental switching fields H_{c180° for all three samples are fitted by the expressions for H_{c2} in Figs. 2(a) to 2(c) and the insets. This way, all switching fields can be nicely fitted by consistently using the same $\epsilon_{90^\circ \pm 2\delta}$ and K_{u1} values for the complete range of angles.

In case of two successive DW nucleations, H_{c4} is not an experimental observable switching field. H_{c4} only indicates $\Delta E_{90^\circ + \delta \rightarrow -\delta} = \epsilon_{90^\circ + 2\delta}$. We have plotted in Figs. 2(a) and 2(b) the “virtual” H_{c4} values for samples A and B. When $0^\circ < \phi < 45^\circ$, the two successive DW nucleations appear as one-step loops for $H_{c2} > H_{c4}$ (see Fig. 3(a)). For $H_{c2} < H_{c4}$, the magnetization loops reveal a two-step behavior with two separate 90° DW nucleations at H_{c2} and H_{c4} , respectively. When $45^\circ < \phi < 90^\circ$ and $K_{u1} > \epsilon_{90^\circ}$, the magnetization switches from $[010]$ to $[\bar{1}00]$ at H_{c1} , where $\Delta E_{270^\circ \rightarrow 180^\circ} = \epsilon_{90^\circ}$. Because $\Delta E_{90^\circ \rightarrow 0^\circ}$ decreases with increasing applied field and becomes ϵ_{90° at H_{c4} (see Fig. 3(b)), two successive DW nucleations appear for $H_{c2} < H_{c4}$. Upon further increasing the field, $\Delta E_{90^\circ \rightarrow 0^\circ}$ becomes negative, and finally reaches $-\epsilon_{90^\circ}$ at H_{c3} , where the magnetization switches backwards from $[100]$ to $[010]$. As a result, the magnetization loops contain three steps. For $H_{c2} > H_{c4}$, the domains aligned along 90° are energetically stable when the applied field exceeds H_{c2} , resulting in two-step loops.

By comparing the expressions for H_{c2} and H_{c4} the field orientation for the occurrence of one-step or three-step loops can be obtained. For the simple case $\delta = 0$, the condition $\tan \phi < K_{u1}/\epsilon_{90^\circ}$ needs to be satisfied, where $0 < \phi < 45^\circ$ for the one-step loops and $45^\circ < \phi < 90^\circ$ for the three-step loops, respectively. Our model predicts that the ranges of angles for which a one-step loop should be observed are $-45^\circ < \phi < 45^\circ$, $-28^\circ < \phi < 28^\circ$, and $-43^\circ < \phi < 36^\circ$ for samples A, B, and C, respectively. The critical angles separating the occurrence of two-step and three-step loops are $\phi = 90^\circ \pm 13^\circ$ for sample A. Our model calculations nicely agree with experiment.

Introducing two successive DW nucleations allows us to consistently interpret the 180° magnetization reorientation. Since the DWs induced during the first nucleation

are energetically unstable, it should be very hard to observe these intermediate domains. In real films, however, DW nucleation and propagation are often perturbed by defects and roughness [24], implying magnetic switching is not as sharp as predicted. As illustrated in Fig. 4(a), we observed the metastable intermediate states in the loops of sample A close to the critical angles separating the occurrence of two-step and three-step loops. The blue curve (increasing field) reveals an overshoot for magnetic

switching from $[\bar{1}00]$ to $[100]$, indicating that the Fe spins align for a short time along $[010]$ before jumping to $[100]$. The red curve (decreasing field) reveals a similar feature. We note that the second intermediate state in the blue curve in Fig. 4(a) is not collinear with the first intermediate state in the red curve and vice versa, which is different from the loop shown in Fig. 1(d). The non-collinearity

implies that not all spins switch from $[\bar{1}00]$ to $[010]$ and then to $[100]$, but some of the spins remain aligned along $[010]$. This points to coexistence of magnetic switching processes with both two-step and three-step loops. The overshoot can still be observed a few degrees away from the critical angles. In sample B we observe a mixture of one-step and two-step loops, as illustrated in Fig. 4(b). The red curve of the transverse MOKE loop reveals two separate 90° DW nucleations, while the blue curve corresponds according to our model to two successive and indistinguishable DW nucleations. Experimentally, the transition between two reversal mechanisms does not occur at one critical angle as predicted by theory, but extends over a small finite range of angles. Time resolved MOKE may be able to detect the ultrafast magnetization dynamics and the intermediate domain formation in the process of two successive DW nucleations.

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FIGURE CAPTIONS

FIG. 1. (Color online) (a) Definition of the angles that are used to describe a film with in-plane cubic anisotropy

and UMA. Typical longitudinal MOKE loops for sample A with (b) one step at $\phi = 8^\circ$, (c) two steps at $\phi = 88^\circ$ and (d) three steps at $\phi = 68^\circ$. The blue (red) curves are for applied fields varying from negative (positive) to positive (negative) saturation. The arrows enclosed by a square represent the orientation of the Fe spins.

FIG. 2. (Color online) The experimental switching fields (symbols) as a function of the field orientation ϕ , and the corresponding theoretical curves for H_{c1} (magenta), H_{c2} (red), H_{c3} (purple), H_{c4} (blue), the “virtual” H_{c4} (dashed gray), and H_c (green) for (a) sample A, (b) sample B, and (c) sample C.

FIG. 3. (Color online) Energy differences $\Delta E_{180^\circ \rightarrow 0^\circ}$ (red), $\Delta E_{270^\circ \rightarrow 180^\circ}$ (cyan), $\Delta E_{180^\circ \rightarrow 90^\circ}$ (green), and $\Delta E_{90^\circ \rightarrow 0^\circ}$ (blue) as a function of the applied field for (a) sample B at $\phi = 10^\circ$ and (b) sample A at $\phi = 65^\circ$.

FIG. 4. (Color online) (a) The longitudinal magnetization loop of sample A at $\phi = 76^\circ$ and (b) the transverse magnetization loop of sample B at $\phi = 29^\circ$ reveal overshoots near the critical angles for the field orientation.







